X-ray powder diffraction under pulsed magnetic fields up to 30T

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P. Frings et al., Rev. Sci. Instr. **77**, 063903 (2006).

Theory: Z. A. Kazeĭ, Moscow State University.

Overview

- Why high magnetic fields?
- How to generate magnetic fields?
- Pulsed magnetic fields
- Application to X-ray diffraction
- Example: Jahn-Teller transition of $\mathrm{TbVO_4}$
- Outlook: Future developments

Why high magnetic fields?

- The magnetic field is a thermodynamic variable of fundamental importance, as temperature or pressure.
- All electrons carry a spin, and therefore a magnetic moment.
  Therefore, in principle, all condensed matter is concerned:

  Magnetically ordered systems (changes of magnetic structure),
  Polymers (orientation),
  Semiconductors (quantum Hall effect),
  Superconductors (flux line lattices, destruction of superconductivity)
  . . . and many others
- The higher the available field,
  the larger the number of phase transitions and other effects
  that can be observed.

How do you generate a magnetic field?

- Up to 1 T: Permanent magnets.
- Up to 15 T: Superconducting magnets → ID20, 10 T.
- Up to 33 T: Resistive magnets, 20 MW.
- Up to 45 T: Hybrid superconducting and resistive magnets, (NHMFL, Tallahassee, 24 MW, ≈ 15 M$).
- → Up to (existing) 80 T: (project) 100 T: Pulsed resistive magnets, ←
- Up to ≈ 130 T: Destructive pulsed magnets (destroys magnet only).
  Up to ≈ 600 T: Destructive pulsed magnets (destroys everything).
- above that: Neutron stars, solar storms, …

Current maximum field for x-ray or neutron diffraction: 15 T (17.5 T with “booster”)

Motivation/Scientific case

- There are many laboratories in Europe and elsewhere in the world which are dedicated to high magnetic field research.
- These labs employ a large number of different techniques:

→ Magnetization and susceptibility.

→ Transport (resistivity, Hall effect, magneto-resistance).

→ Specific heat.

→ Dilatometry and sound velocity.

→ De-Haas-van-Alphen effect (Fermi surface mapping)

→ NMR (Nuclear magnetic resonance)

→ Optical spectroscopy (Raman scattering, reflectivity, ellipsometry, . . .)

Motivation/Scientific case

- All of the current techniques are macroscopic measurements.

. . . but there is no information about the microscopic structure of the sample at fields above 15 T!

- At the same time we know (from measurements at lower fields) that often field-induced phase transitions have a structural component.
- Sound velocity and dilatometry measurements at high fields also indicate structural effects.

There is an urgent need for diffraction for fields above 15 T!
→ Find the easiest and most cost-effective way
to explore this region of the phase diagram. . .

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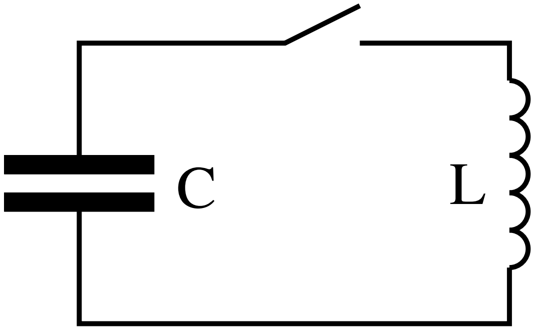
Installations become progressively bigger, more expensive, and more difficult to manage, with exception of pulsed fields, which are scalable.

How to generate pulsed magnetic fields?

The principle is very simple:

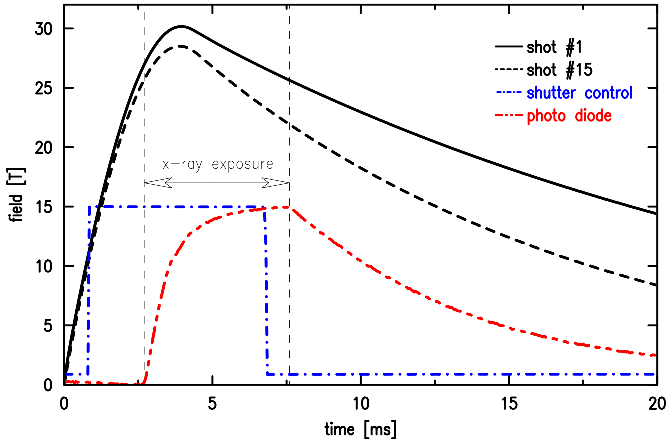


1) Charge capacitor
2) Close switch
3) Current flows
4) . . . repeat

$$\tau = \sqrt{LC}$$

$$E = \frac{1}{2}CV^2 = \frac{1}{2}LI^2$$

$$E = \frac{1}{2\mu_0}\int \mathrm{d}V B^2$$



. . . but some details need to be considered:

- High voltage/high current risks: 24 kV, 6 kA

→ Grounding, protection of beamline electronics . . .

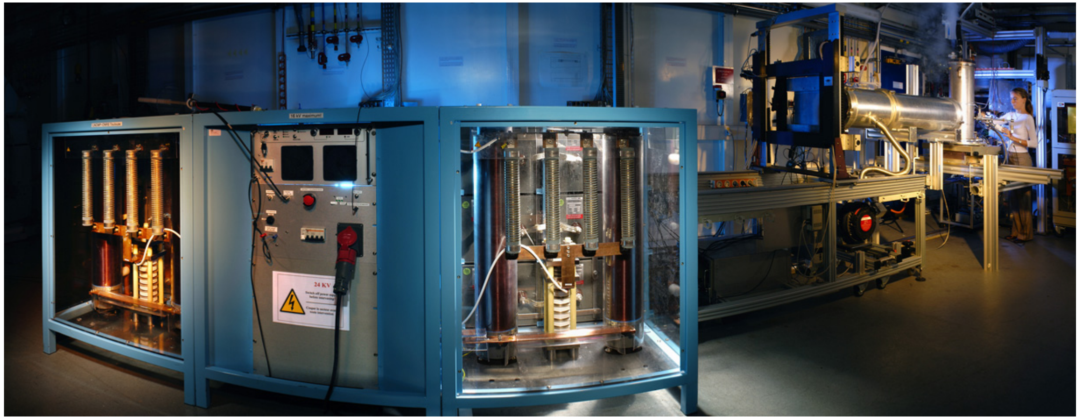
- Stored energy:110 kJ (upgrade to 1.5 MJ planned)

→ transformed into heat at the end of the pulse . . .

→ . . . need efficient cooling of the coil

→ What happens in case of a fault?

High field laboratories know very well how to handle this.



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Application to x-ray diffraction

- Magnet and capacitor bank supplied by LNCMP/Toulouse

→ Transportable capacitor bank, 130 kJ energy, 2.8 tons, $\approx 4\,\mathrm{m}^3$.

→ Solenoid magnet, lq. N_2 cooled, maximum field 30 T, bore 22 mm, max. opening angle 22°.

→ rise time 5 msec, decay time ≈ 20 msec, 10 shots per hour.

- X-ray powder diffraction at 21 keV

→ Online-image plate detector

→ Fast shutter to synchronize the x-ray exposure to the magnetic field pulse.

30 T limit convenient because of wire material → duty cycle, fatigue, . . .
→ upgrade to 60 T relatively straightforward

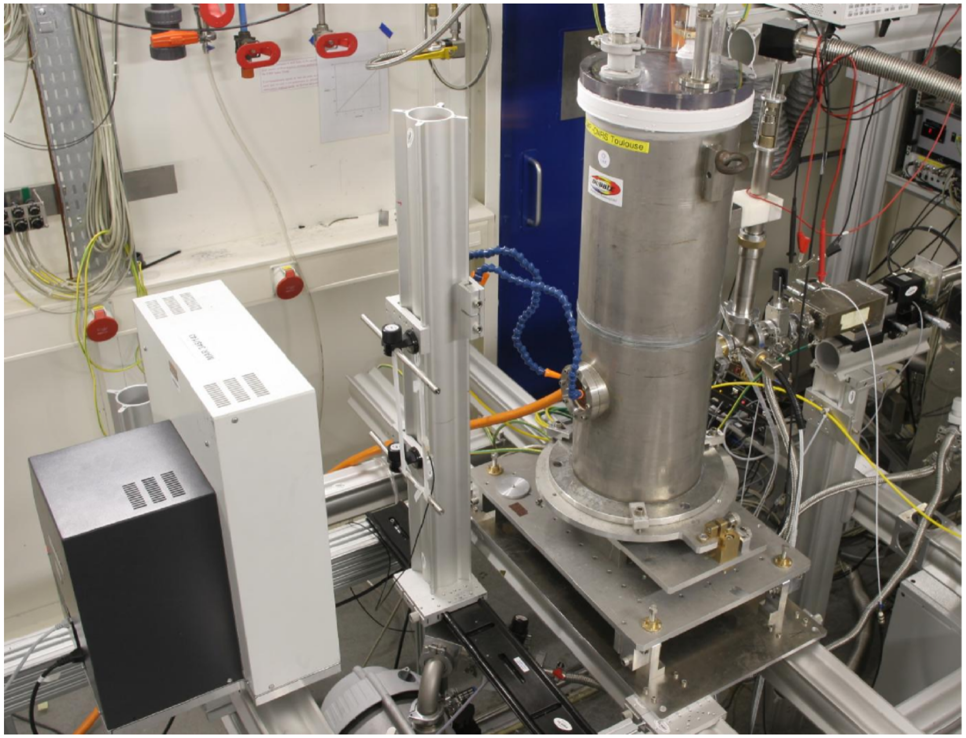
X-ray powder diffraction on BM26B DUBBLE

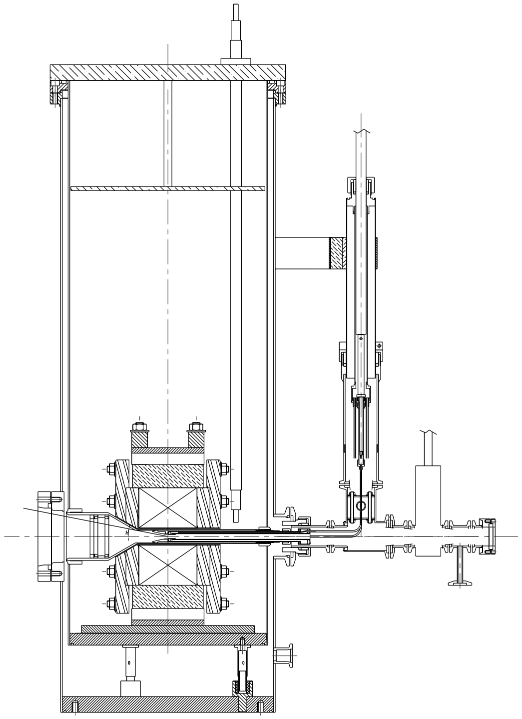
Transportable generator:

- 2 storage modules,
  1 charger/control module
- $C = 1\,\mathrm{mF}$, $V_{\max} = 16\,\mathrm{kV}$, $E_{\max} = 130\,\mathrm{kJ}$
- Total weight $\approx 2.8\,\mathrm{t}$
- Total size $(h \times d \times w)$
  $1.25 \times 1.30 \times 2.85\,\mathrm{m}^3$
- Generator and load magnet installed in radiation hutch.
- Interlocked through radiation hutch PSS.
- Remote control over fiber optical cables.

Generator design: P. Frings (LNCMP).

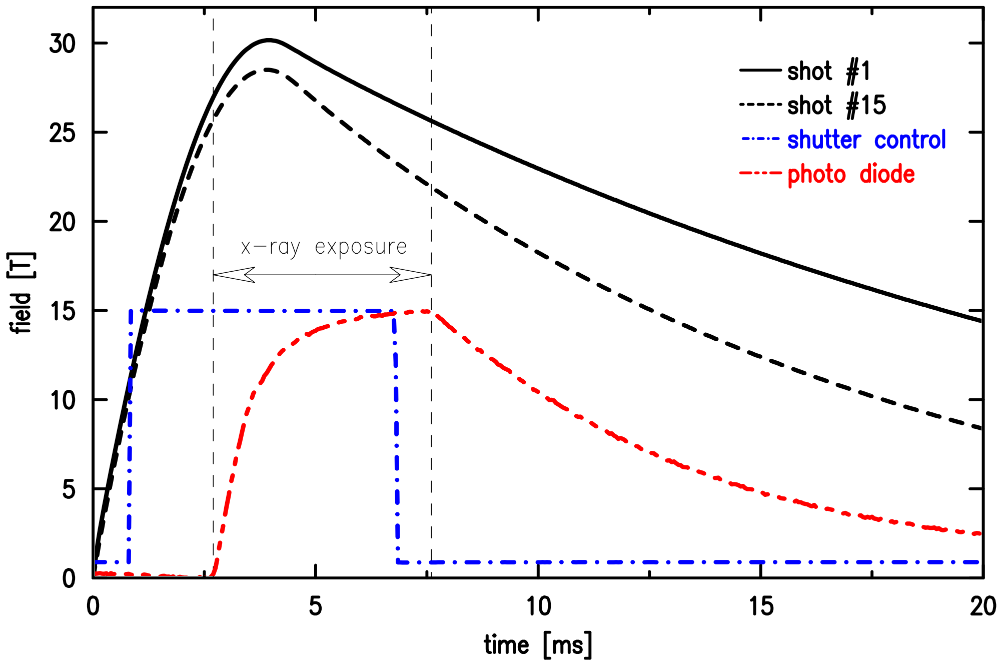
X-ray powder diffraction on BM26B DUBBLE





Coil design: J. Billette (LNCMP), cryostat design: M. Nardone, A. Zitouni (LNCMP).

X-ray powder diffraction on BM26B DUBBLE



- Shutter synchronized to magnetic field pulse
- Warming of coil after sequence of pulses.
- Signal integrated over $\approx 5\,\mathrm{ms}$ per pulse.

Not ultra-fast, but not stroboscopic: Small number of pulses.
Fatigue life: Design system such that 1 shot is enough.

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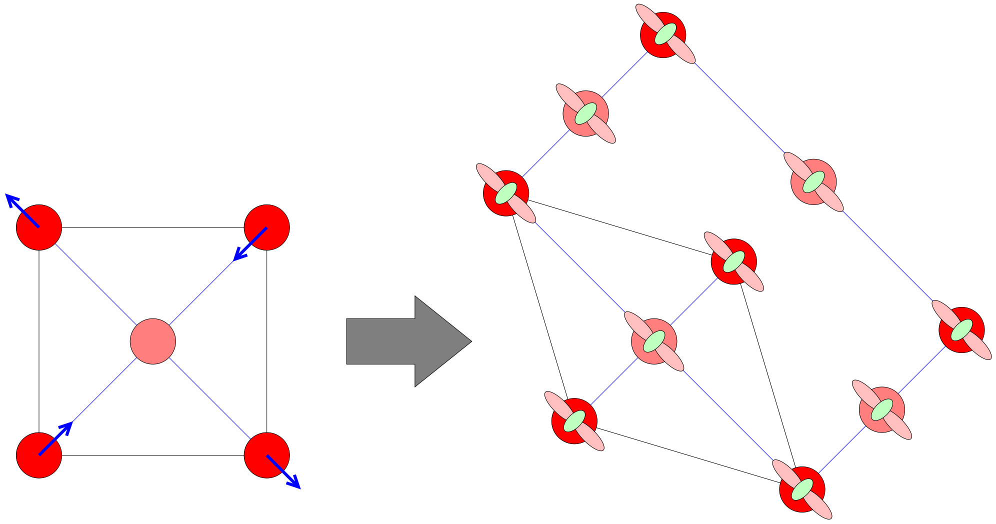
Example: Jahn-Teller transition in $\mathrm{TbVO_4}$

- $\mathrm{TbVO_4}$ is a textbook example of a cooperative Jahn-Teller transition mediated by quadrupolar interactions. $T_{\mathrm{JT}} \approx 34\,\mathrm{K}$.
- The system is known since the 1970'ies and has been studied intensively at zero field. G. A. Gehring and K. A. Gehring, Rep. Prog. Phys. **38**, 1 (1975).

- Driven by $\mathrm{Tb}\ 4f$ quadrupole moment
- Orthorhombic distortion, 2% along (111)
- Space group $I4_1/amd \rightarrow Fddd$

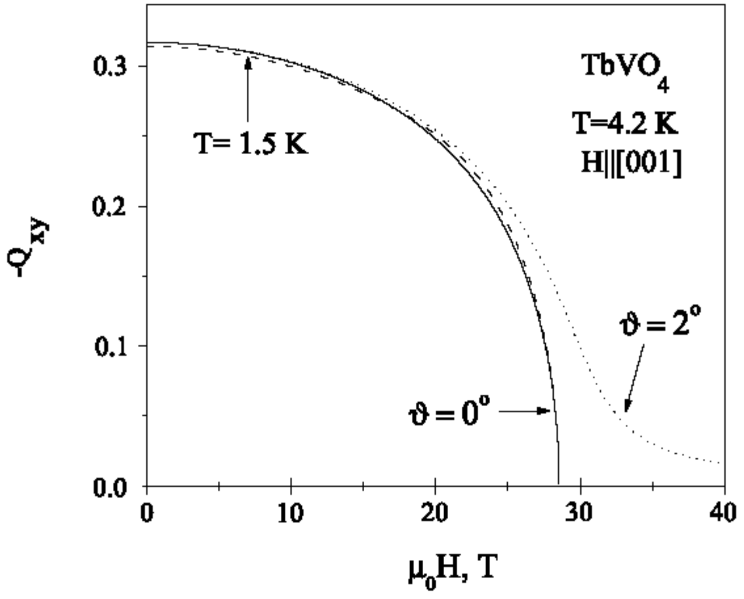


Example: Jahn-Teller transition in $\mathrm{TbVO_4}$

- Recently, first studies in high magnetic fields.
- Strongly anisotropic response (CF).
- Theory (qualitative): 1970'ies
  Competition of magnetization and quadrupolar moment
- Theory (quantitative):
  Field of $\approx 28\,\mathrm{T}$ along the c-axis suppresses the JT state.

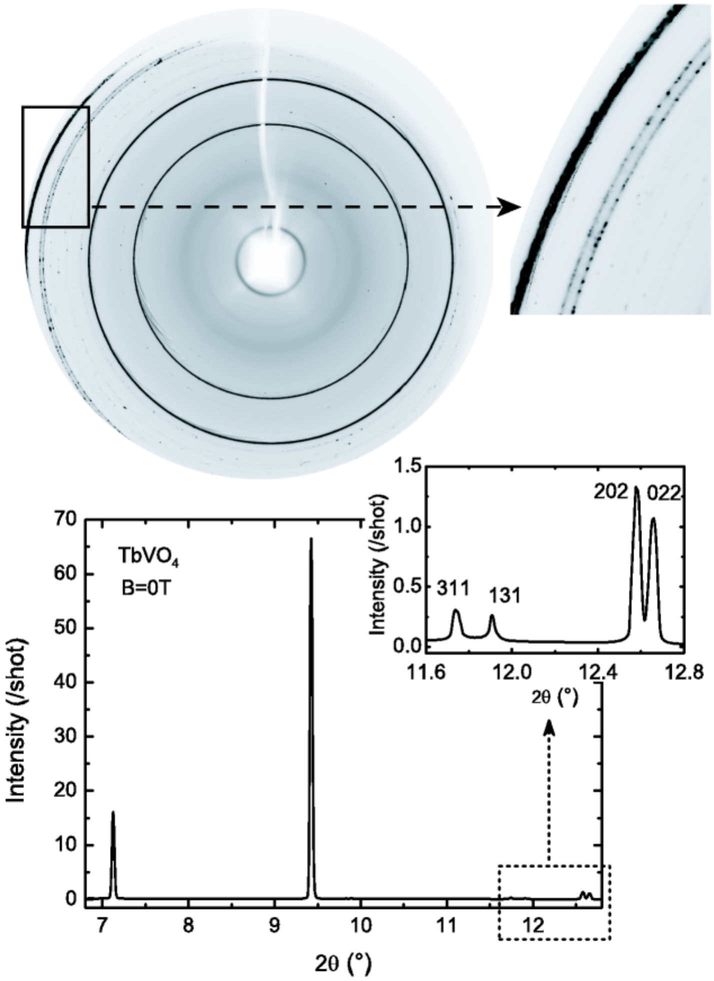
  A. A. Demidov et al, Physica B **363**, 245 (2005).
- Indirectly observed in magnetization.

  Kazei et al, JETP Lett. **82**, 609 (2005).
- But so far no direct observation



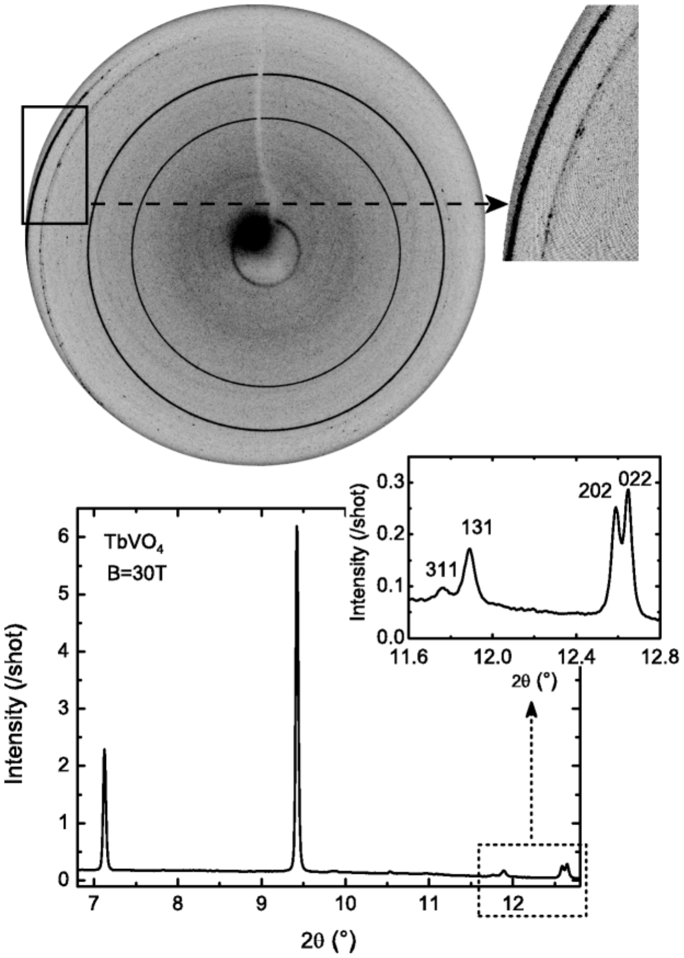
Jahn-Teller transition in $\mathrm{TbVO_4}$: Raw data

- DUBBLE CRG at ESRF
- 21 keV
- MAR 345 image plate detector
- Exposure time $60\,\mathrm{s}$
- $B = 0\,\mathrm{T}$, $T = 7.5\,\mathrm{K}$
- Sample:
  Ground single crystals
  embedded in a polymer matrix
  to suppress grain movement
  and improve thermal contact.

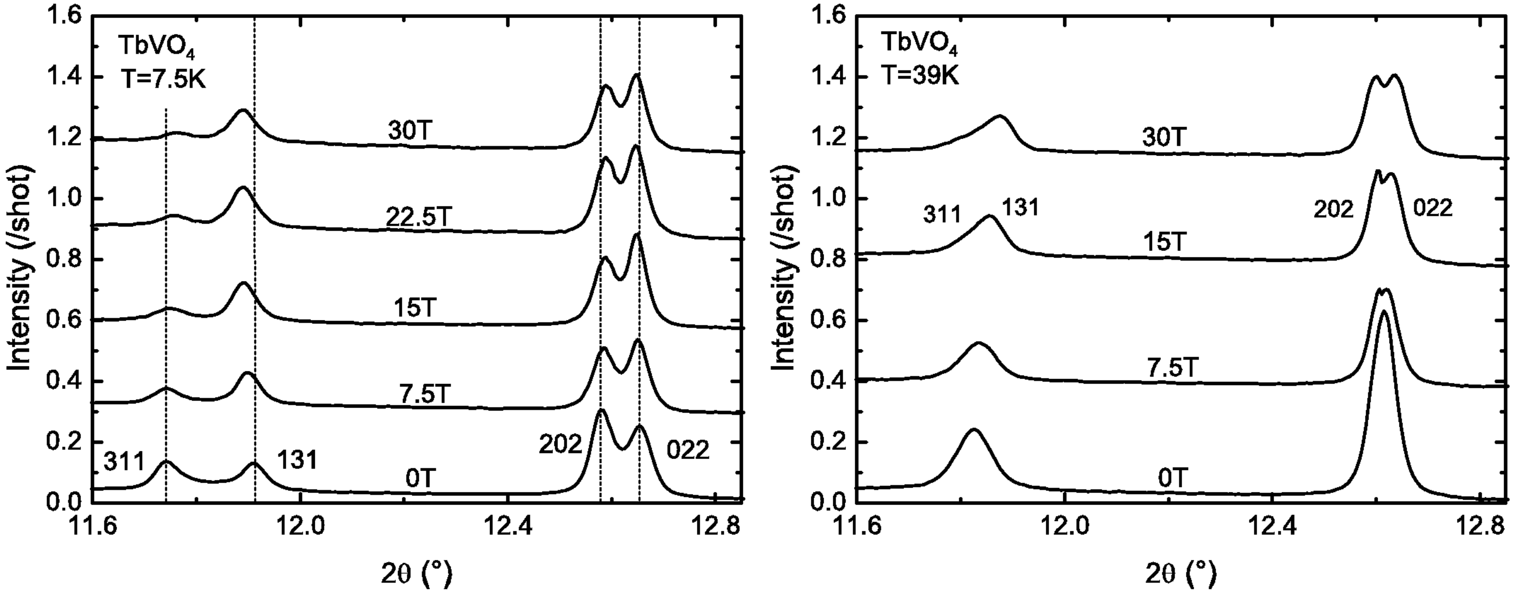


Jahn-Teller transition in $\mathrm{TbVO_4}$: Raw data

- DUBBLE CRG at ESRF
- 21 keV
- MAR 345 image plate detector
- Exposure time $15 \times 5\,\mathrm{ms}$
- $B = 30\,\mathrm{T}$, $T = 7.5\,\mathrm{K}$
- Sample:
  Ground single crystals
  embedded in a polymer matrix
  to suppress grain movement
  and improve thermal contact.



Jahn-Teller transition in $\mathrm{TbVO_4}$: 2θ scans



- High temperature: small splitting induced by magnetic field.
- Low temperature: Splitting reduced by magnetic field.

→ Complex average over phase diagram because of powder average

Interpretation (qualitative)

- The system is driven by the Tb $4f$ quadrupole moment

$$\epsilon \propto Q_{xy} = \frac{1}{2}\left(J_x J_y + J_y J_x\right)$$

Very strong L-S coupling in rare earths
links magnetic moment and charge distribution.

- Coupling between quadrupole and magnetic dipole induced by magnetic field.

→ $B \parallel (001)$: Magnetization $\propto J_z$ in competition with Q_{xy}.

→ $B \parallel (110)$: Magnetization $\propto (J_x + J_y)$ increases Q_{xy}.

- In a powder sample: Average over all possible directions

→ Average over different phase diagrams

- Working on quantitative data analysis with Z. A. Kazeĭ.

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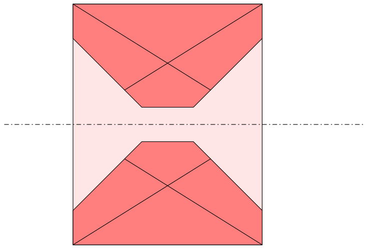
Toulouse 30T magnet system: Second generation

- New coil design for increased optical access (J. Billette, LNCMP)

  → Coil wound onto a double-cone
  → opening angle up to 31°
  → more powder lines available for measurement



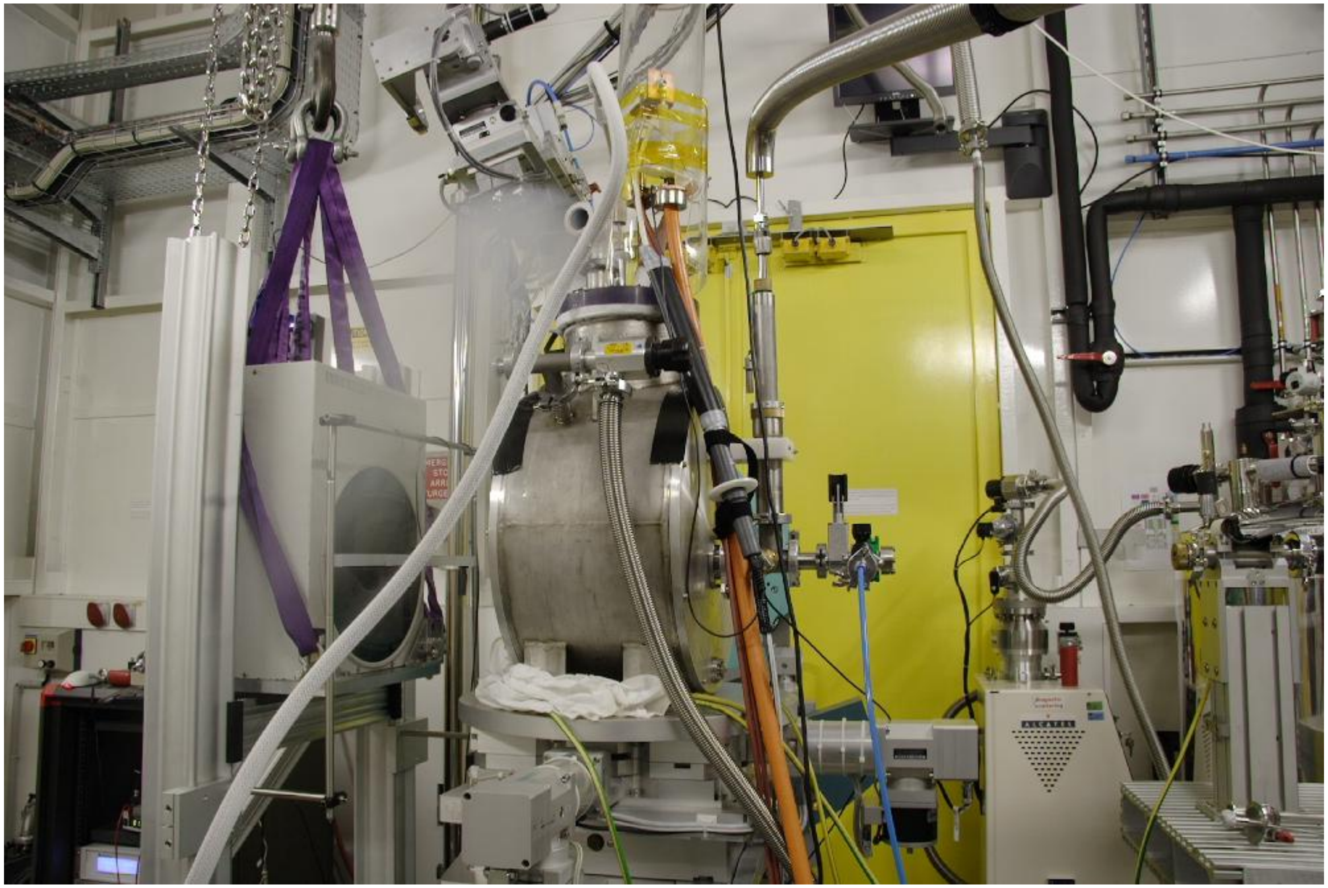
- Installation on undulator beamline ID20

→ $\approx \times 50$ gain in intensity
→ Generator installed outside the radiation hutch

- First tests on the beamline 08–14/11/2006

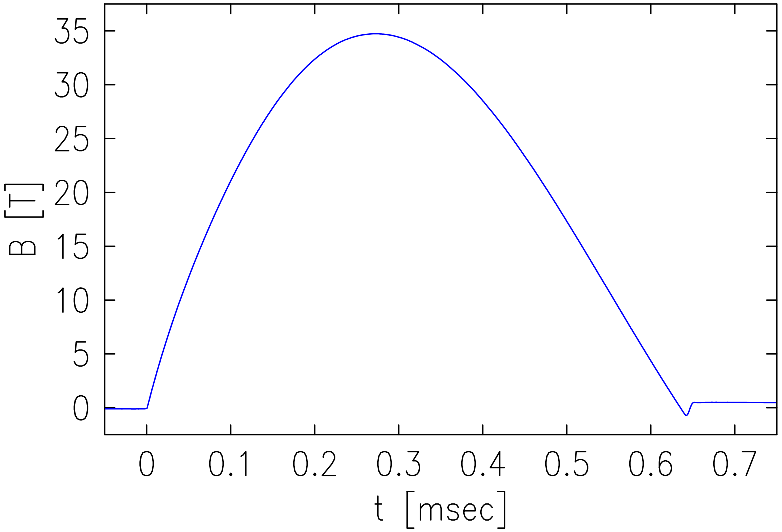
→ Sufficient intensity with 2–4 msec exposure time
→ Need only one shot per spectrum

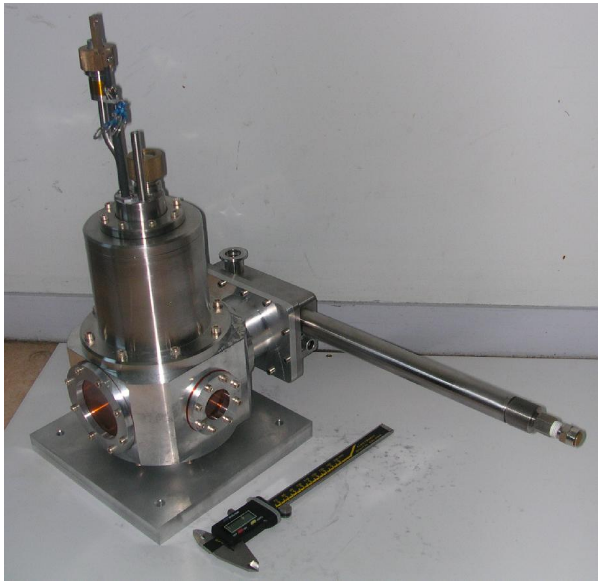
Toulouse 30T magnet system: Second generation



Miniature pulsed magnetic field coils

(Peter van der Linden, Olivier Mathon)



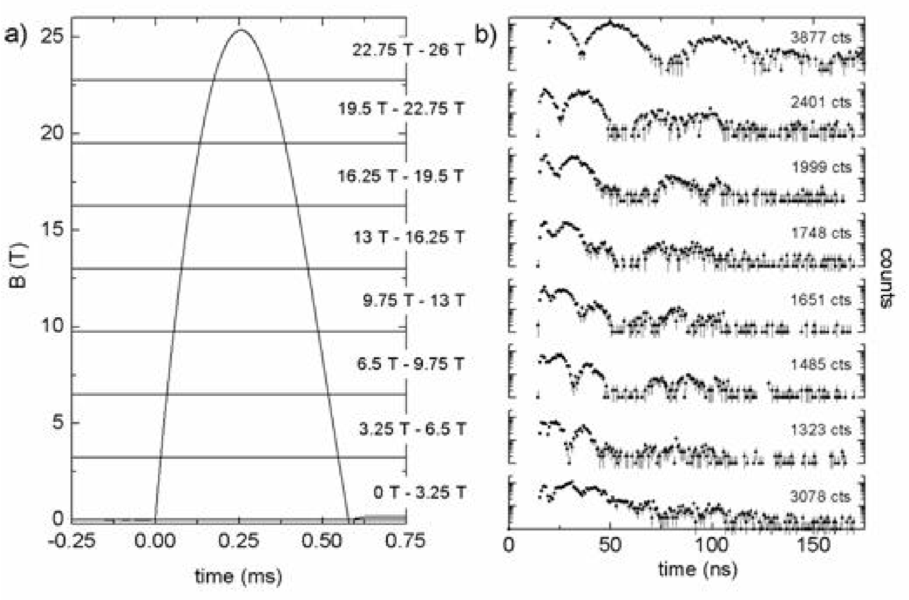


Successfully tested in X-ray magnetic circular dichroism (XMCD) experiments on ID24

- Very compact system, can be installed on any beamline.
- Rise time 250 μs, one pulse every 10 sec.

Nuclear resonant forward scattering using pulsed magnetic fields

(Cornelius Strohm, Peter van der Linden, Rudolf Rüffer)



- Nuclear Resonant Forward Scattering of $^{57}\mathrm{Fe}$ foil
- Using mini-coil system
- Total data acquisition time $\approx 8\,\mathrm{h}$

Future developments

Short term:

→ The technical solution we are using now has a lot of potential.

→ Significant improvements are necessary before this can become a standard experiment with a user program.

→ For most experiments a split coil geometry with $\vec{B} \perp \vec{k}$ is desired.

→ Try other x-ray techniques: Spectroscopy (EXAFS, XMCD), Laue diffraction can be done by installing our equipment on different beamline.

Medium/long term:

→ Need to improve the detection efficiency. Fast 2D pixel detector?

→ Very low temperatures, down to $100\,\mathrm{mK}$.

→ Higher field, up to $60\,\mathrm{T}$. Improved duty cycle of the magnet system.

→ A permanent setup for capacitor banks, optimized detection system, etc.

Summary/Conclusions

- X-ray diffraction under high magnetic fields is virtually virgin ground. There is plenty to be done.
- Steady magnetic fields have the advantage that we can use proven measurement strategies, measure very small signals, etc.
- Pulsed magnetic fields require much more development of x-ray diffraction.
- But because of sample volume, time structure, etc, they can boldly go where no neutron has gone before (and very likely will ever go*).

→ There is a scientific case for both of them.

→ Steady fields solution is lower risk, but limited to 30–40 T.

→ Pulsed fields solution is much more speculative. But it also requires less capital investment, and the ms time resolved x-ray techniques may be of interest in other fields, such as on-line chemistry, shock waves, ...

* . . . with the possible exception of neutron stars!